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# Multiphoton ionization potentials for H and He atoms in intense circularly polarized laser beams

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Intensity—dependent ionization potentials for atomic hydrogen and helium in the presence of intense circularly polarized laser beam are computed using a theory published earlier. The theory employs a unitary transformation which turns out to be identical to the one proposed by Kramers. Results are given both as a function of intensity and frequency of the incident radiation and are compared with those available for the case of plane polarized beam of equivalent intensity. It is found that the circularly polarized radiation is more efficient in multiphoton ionization processes.

## 1. INTRODUCTION

The development of laser as a source of intense radiation has stimulated much interest in the phenomena of breakdown of gases ($\geqslant 10^{-2}$ Torr) due to a Q-switched laser beam (Meyer & Haught 1963, Maker & Terhune 1964, Damon & Tomlinson 1963, Minc 1964). The mechanism responsible for initiating the discharge at optical frequencies is attributed to the process of inverse Bremsstrahung (Wright 1964, Zel'dovich & Raiser 1965, Tozer 1965, Tomlinson 1965). The breakdown process, however, requires for its initiation the existence of free electrons. It has been predicted by Tozer (1965) and more conclusively by Gold & Bebb (1965) that the source for such electrons is the direct ionization of atoms through simultaneous absorption of several photons.

An experiment carried out by Voronov & Delone (1965) at a pressure $< 10^{-2}$ Torr has established that multiphoton effect is inherently nonlinear. More recent experiments (Delone *et al* 1968, Agostini *et al* 1970, Luvan *et al* 1973) have shown that the number of photons, $K$ involved in the ionization process is smaller than $K_0$ obtained from the ionization potential divided by the photon energy $\hbar\omega$. Further, the ionization probability is reported to be proportional to the radiation intensity raised to the power $K$ for $K < K_0$. The fact that smaller number of photons is required is considered to be the result of the effect of intense radiation field on the bound states of the electron in the atom.

Considerable amount of theoretical work on the multiphoton ionization of gases has been reported especially within the framework of time dependent perturbation theory. Keldysh (1965) approached the problem semiclassically, while most other authors (Bebb & Gold 1965, Morton 1967, Chan & Tang 1969, Gontier & Trahin 1968, 1972) have used the quantized form of radiation and treated the atomic system quantum mechanically. Although the perturbation theory has been remarkably successful, its validity for high field intensities ($\geqslant 10^{11}$ watts/cm$^2$) is in doubt; furthermore, none of these workers have taken into account the effect of intense radiation on the initial state of the atom which cannot be ignored.

The present investigation is based on a perturbation method introduced by one of the authors (Henneberger 1968) which yields an effective potential determined by the time average of the combined effects of the Coulomb and electromagnetic fields. It is this effective potential which leads to a shifted spectrum of "dressed states", thus yielding an improvement over the unperturbed states of the atom in the radiation field. The method makes use of a unitary transformation proposed by Kramers (1950) in order to avoid divergences in non-relativistic quantum electrodynamics in the dipole approximation.

A computation of intensity dependent ionization potentials for atomic hydrogen and helium has been published for the case of plane polarization (Choi *et al* 1974). This paper reports the corresponding result for circularly polarized light. A comparison of results obtained in the two cases of polarization confirms the experimental result (Fox *et al* 1971, Kogan *et al* 1971) that circularly polarized light is of greater efficiency in multiphoton ionization processes.

## 2. General Theory

The Hamiltonian of a bound, non-relativistic electron in interaction with the classical radiation field $\boldsymbol{A}$ is, according to the principle of minimal substitution, written as

$$H = \frac{1}{2m}\left(\boldsymbol{P}-\frac{e}{c}\boldsymbol{A}\right)^2+V(\boldsymbol{r}), \qquad \ldots (1)$$

where $V(\boldsymbol{r})$ is the Coulomb potential. On assuming the dipole approximation $\boldsymbol{A}(\boldsymbol{r}, t) \approx \boldsymbol{A}(t)$, the Hamiltonian in the Coulomb gauge becomes

$$H = H_0+H_{int}, \qquad \ldots (2)$$

where

$$H_0 = \frac{\boldsymbol{P}^2}{2m}+V(\boldsymbol{r}), \qquad \ldots (3)$$

$$H_{int} = -\frac{e}{mc}\qquad\frac{e^2}{2mc^2}\boldsymbol{A}^2. \qquad \ldots (4)$$

The spin magnetic moment interaction term is absent because of the dipole approximation.

The method introduced by Henneberger (1968) consists of performing a time dependent unitary transformation $\Omega(t)$ on the Schrödinger wave function $\psi(\boldsymbol{r}, t)$ so that the new wave function becomes

$$\psi(\boldsymbol{r}, t) = \Omega(t)\psi(\boldsymbol{r}, t), \qquad \ldots \quad (5)$$

with the transformation being defined as

$$\Omega(t) = \exp\left[\frac{i}{\hbar}\int_{-\infty}^{t} H_{int}(t')dt'\right]. \qquad \ldots \quad (6)$$

$\Omega(t)$ may further be written as

$$\Omega(t) = \Omega_1(t)\Omega_2(t), \qquad \ldots \quad (7)$$

where

$$\Omega_1(t) = \exp\left[\frac{i}{\hbar}\boldsymbol{\alpha}.\boldsymbol{P}\right], \qquad \ldots \quad (8)$$

and

$$\Omega_2(t) = \exp\left[\frac{i}{\hbar}\int_{-\infty}^{t}\frac{e^2}{2mc^2}\boldsymbol{A}^2(t')dt'\right]. \qquad \ldots \quad (9)$$

The quantity $\boldsymbol{\alpha}$ is given by

$$\boldsymbol{\alpha}(t) = -\frac{e}{mc}\int_{-\infty}^{t}\boldsymbol{A}(t')dt' \qquad \ldots \quad (10)$$

which, because of the relation

$$\ddot{\boldsymbol{\alpha}}(t) = -\frac{e}{mc}\dot{\boldsymbol{A}}(t) = \frac{e}{m}\boldsymbol{E}(t), \qquad \ldots \quad (11)$$

turns out to be the classical displacement of a free electron from its centre of oscillation in a radiation field.

The transformation (6) yields a new Hamiltonian

$$\mathcal{H} = \Omega(t)H\Omega^{-1}(t) + i\hbar\dot{\Omega}(t)\Omega^{-1}(t), \qquad \ldots \quad (12)$$

which simplies to

$$\mathcal{H} = \frac{\boldsymbol{P}^2}{2m} + V(\boldsymbol{r}+\boldsymbol{\alpha}) \qquad \ldots \quad (13)$$

satisfying the wave equation

$$\left[\frac{\boldsymbol{P}^2}{2m}+V(\boldsymbol{r}+\boldsymbol{\alpha})\right]\psi(\boldsymbol{r},t)=i\hbar\frac{\partial\psi(\boldsymbol{r},t)}{\partial t}. \qquad \ldots (14)$$

Thus we see that the transformation couples the Coulomb and the radiation fields. For no binding energy $V(\boldsymbol{r})=0$, the solution of eq. (14) represents a free particle interacting with the external radiation. Hence the modified Coulomb potential which is now time dependent is

$$V(\boldsymbol{r}+\boldsymbol{\alpha})=-\frac{e^2}{|\boldsymbol{r}+\boldsymbol{\alpha}|}.$$

By making use of the three—dimensional Fourier integeral representation of $\frac{1}{|\boldsymbol{r}+\boldsymbol{\alpha}|}$ the above expression can be written in a more suitable form

$$V(\boldsymbol{r}+\boldsymbol{\alpha})=-\frac{e^2}{2\pi^2}\int\frac{d^3q}{q^2}\exp[i\boldsymbol{q}.(\boldsymbol{r}+\boldsymbol{\alpha})].$$

## 3. Specialization to Circularly Polarized Radiation

The classical displacement of the free electron in the presence of a circularly polarized radiation of frequency $\omega$ may be written as

$$\boldsymbol{\alpha}(t)=\boldsymbol{\alpha}_{10}\sin\omega t+\boldsymbol{\alpha}_{20}\cos\omega t, \qquad \ldots (15)$$

where $\boldsymbol{\alpha}_{10}=\alpha_0\boldsymbol{\epsilon}_1$, and $\boldsymbol{\alpha}_{20}=\alpha_0\boldsymbol{\epsilon}_2$.

In the above equations, $\boldsymbol{\epsilon}_1$ and $\boldsymbol{\epsilon}_2$ are unit vectors along the $X$ and $Y$ axes respectively, and $\alpha_0$ is the amplitude of displacement. Eq. (15) yields

$$e^{i\boldsymbol{q}.\boldsymbol{\alpha}}=\sum_{p=-\infty}^{\infty}\sum_{s=-\infty}^{\infty}J_p(\boldsymbol{q}.\boldsymbol{\alpha}_{10})J_s(\boldsymbol{q}.\boldsymbol{\alpha}_{20})(e^{i\omega t})^{p+s}(e^{i\pi/2})^s. \qquad \ldots (16)$$

where $J_p$ and $J_s$ are Bessel functions of order $p$ and $s$ respectively. $p$ and $s$ are integers with both positive and negative values.

To study the intensity dependent ionization energy, one includes in the Hamiltonian time independent part of $V(\boldsymbol{r}+\boldsymbol{\alpha})$. Therefore, on setting $p+s=0$ in eq. (16), one obtains after some manipulation the effective potential as seen by the electron :

$$V_0(\boldsymbol{r})=-\frac{e^2}{2\pi^2}\int\frac{d^3q}{q^2}e^{i\boldsymbol{q}.\boldsymbol{r}}J_0\left(\alpha_0\sqrt{(\boldsymbol{q}.\boldsymbol{\epsilon}_1)^2+(\boldsymbol{q}.\boldsymbol{\epsilon}_2)^2}.\right) \qquad \ldots (17)$$

Clearly this potential now depends on both the Coulomb and the radiation fields. It is of interest to see what charge distribution actually produces such an effective

potential. By means of Poisson's equation it is seen that the effective charge density in question is given by

$$\rho_{eff}(\boldsymbol{r}) = \frac{e}{8\pi^3} \int d^3q\, e^{i\,\boldsymbol{q}\cdot\boldsymbol{r}}\, J_0\left(\alpha_0\sqrt{(\boldsymbol{q}\cdot\boldsymbol{\epsilon}_1)^2+(\boldsymbol{q}\cdot\boldsymbol{\epsilon}_2)^2}\right). \quad \ldots (18)$$

Assuming $\phi_q$ and $\phi$ to be the azimuthal angles with respect to the coordinate axes $(XYZ)$ of $\boldsymbol{q}$ and $\boldsymbol{r}$ respectively, eq. (18) can be simplified to

$$\rho_{eff}(\boldsymbol{r}) = \frac{e}{4\pi^2}\,\delta(Z) \int e^{iq_\rho\rho\cos(\phi-\phi)} J_0(\alpha_0 q_\rho) q_\rho dq_\rho d\phi_q, \quad \ldots (19)$$

where the $Z$-integeration has already been performed. After the angular integeration one obtains

$$\rho_{eff}(\boldsymbol{r}) = \frac{e}{2\pi}\,\delta(Z) \int_0^\infty J_0(\alpha_0 q_\rho) J_0(\rho q_\rho) q_\rho dq_\rho. \quad \ldots (20)$$

Eq. (20) is the well known Fourier-Bessel integral (Watson 1966). Thus the effective charge density is found to be

$$\rho_{eff}(\boldsymbol{r}) = \frac{e}{2\pi\rho}\,\delta(\alpha_0-\rho)\delta(Z), \quad \ldots (21)$$

where $e$ is the proton charge. Eq. (21) shows that the effective potential of a bound electron in the field of a circularly polarized radiation is the one produced by a ring distribution of charge $e$ which in the case of hydrogen is that of proton. The radius of the ring is the amplitude $\alpha_0$ of classical displacement of the electron.

The above conclusion immediately helps one write down the potential at a point $(r, \theta)$ (Weber 1950) due to a ring (lying in the $X$–$Y$ plane) of nuclear charge $e$ of atomic hydrogen :

$$V(r,\theta) = \begin{cases} e\sum_{l=0}^{\infty} \dfrac{r^l}{\alpha_0^{l+1}} P_l(\cos\pi/2)\, P_l(\cos\theta), & \alpha_0 > r \\ e\sum_{l=0}^{\infty} \dfrac{\alpha_0^{\,l}}{r^{l+1}}\, P_l(\cos\pi/2)\, P_l(\cos\theta), & r > \alpha_0 \end{cases} \quad \ldots (22)$$

with $V_0(r) = -eV(r,\theta)$. One can similarly write down the potential $V(r_l, \theta_l)$ at the positions of each electron in the case of atomic helium. The Hamiltonians for the atomic systems under investigation are then

$$\mathcal{H}_0^H = -\frac{\hbar^2}{2m}\nabla^2 + V_0(r),$$

and

$$\mathcal{H}_0^{He} = -\frac{\hbar^2}{2m}\sum_{i=1}^{2}\nabla_i^2 - e\sum_{i=1}^{2}V(r_i,\theta_i) + \frac{e^2}{r_{12}}. \quad \ldots (23)$$

## 4. Solution of the Schrödinger Equation Using Basis Functions

The problem now is to solve the time independent Schrödinger equation

$$\mathcal{H}_0\psi = E_0\psi \quad \ldots (24)$$

for the ground state energy $E_0$ of the atom under consideration. The energy corresponds to the ionization potential and is a function of intensity of the field. $\mathcal{H}_0$ is the appropriate Hamiltonian given by eq. (23).

As the potential is of complicated nature, the Schrödinger equation cannot be solved exactly. One must, therefore, resort to some method of approximation. Since the primary interest is to obtain an approximate estimate of $E_0$, the variational method turns out to be an excellent approximation. According to this method the eigenfunction $\psi$ is approximated by a trial wave function $\phi$ so that

$$<\mathcal{H}_0> = \frac{\int \phi^* \mathcal{H}_0 \phi d\tau}{\int \phi^* \phi d\tau} \geqslant E_0. \quad \ldots (25)$$

The trial wave functions are of the form

$$\phi = \sum_{n=0}^{N}\sum_{l=0}^{n} C_{n,l}\chi_{n,l}(\mathbf{r}),$$

with

$$\chi_{n,l}(\mathbf{r}) \equiv r^n e^{-\beta r} P_l(\cos\theta). \quad \ldots (26)$$

Corresponding symmetrized trial functions have been used for the helium calculations. It is unnecessary to assume a dependence of the $\chi_{n,l}$ on azimuthal angle, since only the ground state is needed for the lowest order calculations. $N$ determines the number of basis functions. In principle it should extend to infinity but the hope is that the parameter $\beta$ which has been incorporated in the exponential will, for its optimum value, provide sufficiently fast convergence of the eigenvalues so that termination occurs after a finite number of basis functions. The variation method leads to the secular equation for atomic hydrogen,

$$|<\chi_{n_i,l_i}|\mathcal{H}_0^H - E_0|\chi_{n_j,l_j}>| = 0. \quad \ldots (27)$$

One similarly obtains the secular determinantal equation for atomic helium. The matrix elements

$$\mathcal{H}_{0ij} = <\chi_{n_i,l_i}|\mathcal{H}_0|\chi_{n_j,l_j}>$$

and

$$S_{ij} = < \chi_{n_i, l_i} | \chi_{n_j, l_j} >$$

have been calculated analytically for both the cases of atomic hydrogen and helium. In case of hydrogen, a set of 30 basis functions corresponding to $0 \leqslant n \leqslant 9$, and $0 \leqslant l \leqslant n$, with $l$ always even were used. For the helium calculations, a set of 36 basis functions were used. At extremely small intensity ($\alpha_0 = 10^{-4}$ a.u.), the computed ionization potentials for atomic hydrogen and helium agreed with known values (no external field) to four significant figures.

The results of the ionization potentials are given in graphical form, together with the results of Choi *et al* (1974) for the plane polarized case (in figures 1–3). The abscissa are $\alpha_0$ for the plane polarized case, and $\sqrt{2}\alpha_0$ for the circularly polarized case. For a given frequency, a value of the abscissa corresponds to the same intensity for plane or circular polarization.

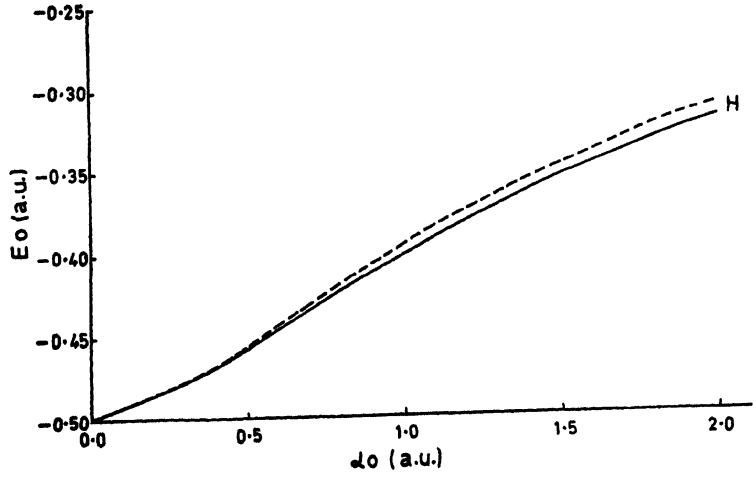


Fig. 1. The ground state energy, $E_0$ of atomic hydrogen versus the classical displacement, $\alpha_0$ of Choi. Solid line is obtained by Choi and the dashed line according to the present computation for equivalent intensity.

## 5. Conclusion

The purpose of this paper is merely to demonstrate that as in the plane polarized case, the ionization potential of atoms in presence of circularly polarized radiation also differs strongly from the usual ground state energy. The figures 1, 2 and 3 show that at decreasing intensities the ionization energies of hydrogen and helium become closer to the corresponding values obtained in the case of plane polarization. Thus in such intensity regions the ionization energies are independent of the nature of polarization of the laser beam. At higher intensities

the energy curves lie somewhat above those reported by Choi *et al* (1974). The relatively small difference in the ionization energies is, in fact, surprising in view of the completely different nature of the effective potential in the two cases. Our conclusion, therefore, that circularly polarized light is more efficient in

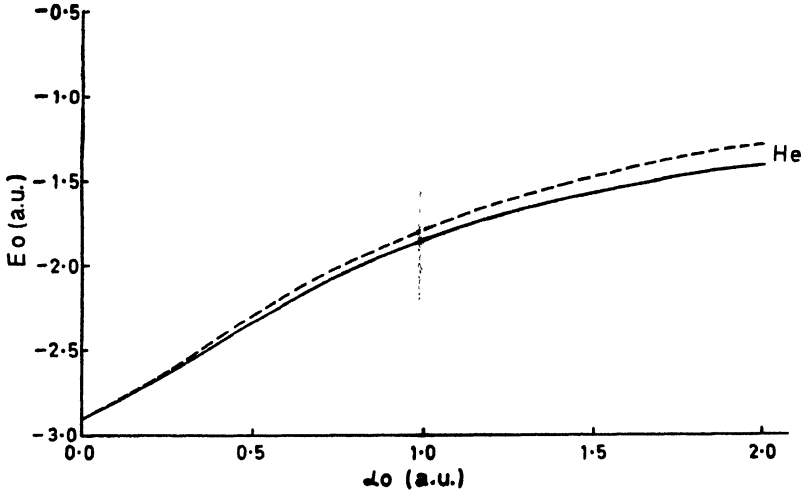


Fig. 2. The ground state energy, $E_0$ of atomic helium versus the classical displacement, $\alpha_0$ of Choi. Solid line is obtained by Choi and the dashed line according to the present computation for equivalent intensity.

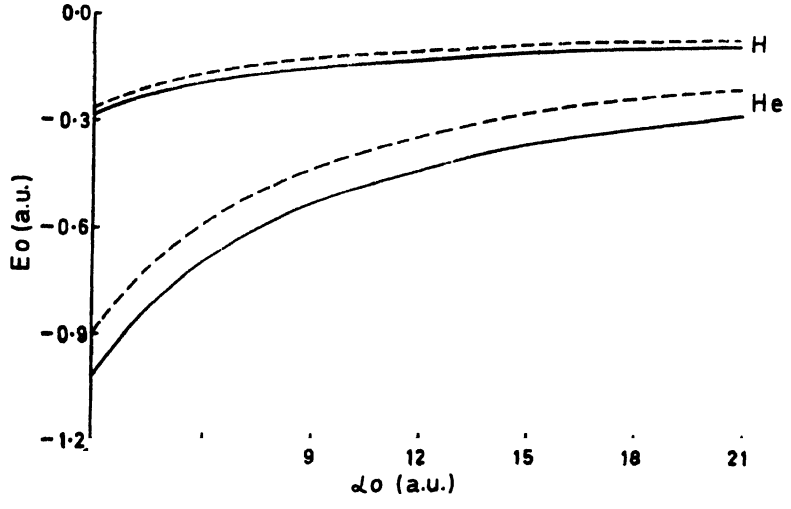


Fig. 3. The ground state energies, $E_0$ of both atomic hydrogen and helium versus the classical displacement, $\alpha_0$. Solid lines are obtained by Choi and the dashed lines obtained according to the present computation for equivalent intensity.

multiphoton ionization than the plane polarized one is consistent with the theoretical predictions of Klarsfeld & Maquet (1971), Faisal (1972), and Lambropoulos

(1972). Since there are no relevant data available, direct comparison of our results with those obtained experimentally is not possible. However, the conclusion also remains consistent qualitatively with the experimental results reported by Kogan *et al* (1971) showing the greater efficiency of circularly polarized light in multiphoton ionization of cesium atoms than the plane polarized one.

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